

Nanoscale Dynamical Mean-Field Theory for molecules and mesoscopic devices in the strong correlation regime

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We develop a nanoscale dynamical mean field theory (nano-DMFT) to deal with strong Coulomb interaction effects in physical systems that are intermediate in size between atoms and bulk materials, taking into account the tunneling into nearby electrodes. Focusing on a simplified tree-like geometry, the usual DMFT loop simply stops when the finite lattice is fully covered, starting with an initial seed provided by the electronic environment at the boundary. To illustrate this nano-DMFT, we investigate the disappearance of the quasi-particle weight in a correlated nano-object near the Mott transition. In contrast to thermally-driven classical phase transitions, quantum effects lead to unexpected oscillations of the order parameter, related to the interference of coherent renormalized quasiparticles. This behavior also implies a spatially inhomogeneous Mott localization process at the nanoscale.

The recent developments in the field of strongly correlated electronic systems are reaching today an interesting point of convergence. Coming from a macroscopic description of bulk condensed matter, powerful many body techniques [1, 2], possibly in combination with ab-initio methods, have been tailored to understand an ever growing number of new compounds with surprising properties [3]. From a bottom-up perspective, the recent experimental progresses in designing artificial atoms (quantum dots) or manipulating single atoms and molecules that are probed by an electronic environment (electrodes) have been met with as well an impressive success [4].

How those two fields will merge together is a challenging question raised by today's research, whether in semiconducting devices with more complex lithographic patterns, or by the study of ultrasmall chunks of correlated materials in quantum transport experiments. Such devices ask however for the development of new theoretical methods as well, since the inclusion of correlation effects in transport calculations by modern numerical methods can only be achieved at the level of single atomic impurities, using Wilson's Numerical Renormalization Group method (NRG) [5, 6], or simplified linear "molecules", by the Density Matrix Renormalization Group [7, 8], but fails for higher-coordination entities.

In the case of correlated materials in the thermodynamic limit, a philosophy based on a *local effective* description has proved nevertheless very successful. Indeed the so-called dynamical mean field theory (DMFT) exactly maps (in the large coordination limit) a macroscopic lattice problem of correlated fermions onto a single atomic level that feels the average effect of the other electrons moving nearby [1, 9]. In this case, the above-mentioned numerical techniques can be used to solve this simplified effective problem, and allow to capture quantitatively the effect of the Coulomb interaction on the low energy electronic properties: strong signatures indeed appear in the electrical conductivity [11, 12] as electrons start to localize near the Mott insulating state [1].

The purpose of this paper is to develop a nano-DMFT

approach for correlated nano-objects which are intermediate in size between the atom and the solid. In the simplest case of tree-like structures, this can be easily formulated using the cavity construction [1], and can be simply described by a standard self-consistency DMFT loop which is stopped at a *finite* number of iterations, related to the linear size of the nano object. This new insight will be applied both to the Ising and Hubbard models respectively, allowing to underline some important physical differences in finite-size many-body effects between the classical and the quantum world. As an illustration for the latter, we will examine how low energy properties cross over from the single atomic impurity to the correlated solid as a function of the system size, focusing on the local quasi-particle weight, a quantity that characterizes the proportion of electrons that survive the Coulomb repulsion at low energy, and can serve as an order parameter for the Mott transition. We find out a surprising oscillatory behavior of this quasiparticle residue as a function of physical parameters, which reveals the interference of coherent quasiparticles in finite size systems. The Mott localization also happens to be non homogeneous at the mesoscale, with a proportion of sites considerably "hotter" than the others (in the sense of scattering properties).

We begin by a general description of the cavity method for finite size lattices in the large coordination limit, see figure 1. Although a derivation of the DMFT equations can be done in principle for an arbitrary geometry of the nano-object, we will consider for simplicity a Bethe lattice of linear size L (determined from its center) and coordination z ; this corresponds to the upper right panel of figure 1 for $L = 2$ and $z = 4$. Focusing on the properties at the center of the lattice (site 0), these can be computed from an effective local action involving the site 0 only, provided one can obtain the dynamics at the nearby sites 1 in the lattice where the site 0 has been removed (cavity), see the lower right panel of figure 1. This statement is exact in the limit where the coordination becomes large [9, 10], but can be taken as a local mean field ansatz

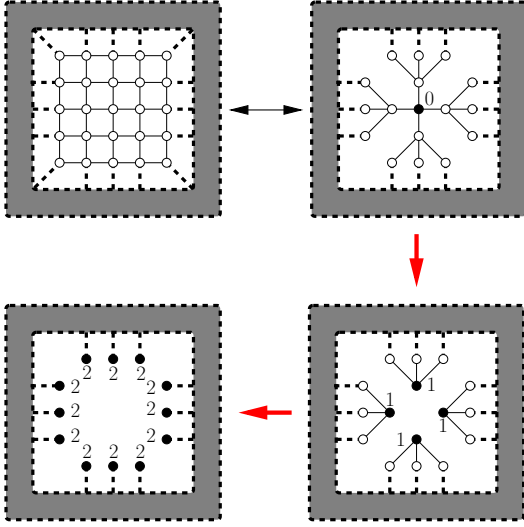


FIG. 1: Correlated nano object of linear extension $2L + 1 = 5$ and coordination number $z = 4$, defined by a finite piece of either the square lattice (top left) or the Bethe lattice (top right). For the latter, which allows the simplest calculations, the nano-DMFT procedure is outlined by the two further stages of a cavity method given in the lower pictures. The external environment, to which the nano object couples, is pictured by the shaded area. The nano-DMFT becomes exact in the limit $z \rightarrow \infty$ keeping L finite.

otherwise. Similarly, the next shell can also be tackled using a action local to the sites 1, given by an effective field provided by the sites 2 (in the cavity where the sites of type 0 and 1 have been removed). This is repeated recursively until the boundary is reached, which amounts to an impurity coupled to a *given* external environment (lower left panel of figure 1).

Let us put the reasoning backwards, focusing for the sake of clarity on the simple Ising model $H = -J \sum_{i,j} S_i S_j - \sum_i h_i S_i$, with a local external magnetic field that couples to the outer shell, $h_L = h \neq 0$ only (due to the tree structure of the lattice, all those sites are independent, so that an extra label is not required). These boundary spins are thus free, i.e. $H^{(L)} = -h S_L$, which gives the thermal average $\langle S_L \rangle = \tanh(h/T)$. This corresponds to the lower left picture in figure 1 for $L = 2$. Now the physics of the sites $L - 1$ can be obtained by an effective impurity problem that reads $H^{(L-1)} = -J \langle S_L \rangle S_{L-1}$ (this is exact as $z \rightarrow \infty$), giving $\langle S_{L-1} \rangle = \tanh(J \langle S_L \rangle / T)$. This equation can be used recursively, until the center (site 0) is reached, providing the results seen on figure 2. Besides the expected monotonous disappearance of the magnetization $\langle S_0 \rangle$ at large system size near the critical temperature $T_c/J = 1$, we remark that the thermodynamic limit is only obtained for rather large system size (which increases with decreasing external magnetic field at the boundary).

We now turn to the more interesting quantum case,

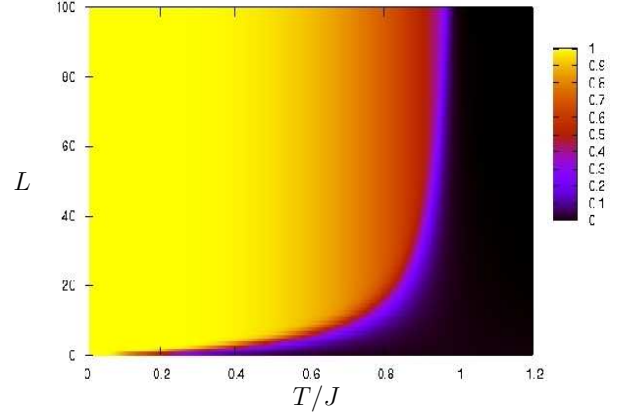


FIG. 2: Average magnetization $\langle S_0 \rangle$ at the center site for a small boundary field $h/J = 0.01$ as a function of temperature T/J (abscissa) and number of sites L (ordinate).

considering a Hubbard-Anderson model [8] for electrons

$$H = -t \sum_{ij,\sigma} d_{i\sigma}^\dagger d_{j\sigma} - \mu \sum_{i\sigma} d_{i\sigma}^\dagger d_{i\sigma} + U \sum_i d_{i\uparrow}^\dagger d_{i\uparrow} d_{i\downarrow}^\dagger d_{i\downarrow} + V \sum_{k\sigma} [d_{L\sigma}^\dagger c_{k\sigma} + \text{h.c.}] + \sum_{k\sigma} \epsilon_k c_{k\sigma}^\dagger c_{k\sigma} \quad (1)$$

with hopping t , chemical potential μ , local Coulomb repulsion U within the correlated lattice, while the hybridization V couples only the outer shell of sites L (an unnecessary extra index labelling the equivalent sites of type L has been omitted above) to an external electronic bath, which models the electrodes in a transport experiment. Following the previous philosophy, we start at the boundary (sites L) with single Anderson impurities (decoupled from the other sites with $i < L$), so that the local Green function $G^{(L)}(\omega)$ is obtained by solving a local effective action which is nothing but the single impurity Anderson model:

$$\mathcal{S}^{(L)} = \frac{1}{\beta} \sum_{n\sigma} [-i\omega_n - \mu + \Delta^{(L)}(i\omega_n)] d_{L\sigma}^\dagger d_{L\sigma} + \int_0^\beta d\tau U d_{L\uparrow}^\dagger d_{L\uparrow} d_{L\downarrow}^\dagger d_{L\downarrow} \quad (2)$$

$$\Delta^{(L)}(\omega_n) = \sum_k \frac{V^2}{i\omega_n - \epsilon_k} \quad (3)$$

where $\beta = 1/T$, $\omega_n = (2n + 1)\pi T$. Equation (3) provides thus the boundary condition (and *not* an effective Weiss field at this first stage), similar to the magnetic field h applied at the boundary in the Ising model. Note that in the absence of an external electronic environment, $\Delta^{(L)} = 0$, the boundary problem reduces to an atomic limit. The nano-DMFT for the full isolated molecule could then allow the calculation of its many-body level spectrum.

Now clearly the effective local action of the sites $L - 1$ takes a form similar to (2), with the simple effective bath:

$$\Delta^{(L-1)}(\omega_n) = t^2 G^{(L)}(i\omega_n) \quad (4)$$

due to the Bethe lattice structure (this is obtained by integrating exactly the sites L in a cavity system constituted of sites $L - 1$ and L only, see the lower right panel of figure 1). By repeating this procedure up to the center of the lattice, we see that *the local Weiss field Green function $\Delta^{(0)}$ at the center site 0 in the Bethe lattice of linear size $2L + 1$ is determined by the L^{th} DMFT iteration loop with an initial condition (3) given by the external electronic environment.*

We now want to investigate some physical results that can be obtained from this nano-DMFT approach. For simplicity, we focus on the case where the probing electrodes are modelled by a constant density of states ρ_0 , so that the external environment is characterized by a single width parameter $\Gamma = \pi\rho_0 V^2$, providing the starting Weiss field of the DMFT loop $\Delta^{(L)}(\omega_n) = -i\Gamma \text{sign}(\omega_n)$. This is then iterated up to a fixed number L of times, corresponding to a given linear size L of the lattice, *for each value* of the Coulomb interaction U and temperature T (the chemical potential is chosen as $\mu = U/2$ to stay at particle-hole symmetry). In contrast, the usual DMFT procedure for a macroscopic system is done until a small and fixed convergence threshold is reached (including a weighting procedure that speeds up this convergence), and does not depend on the boundary condition (unless one considers the first-order metal/insulator transition regime where two solutions coexist, see below).

Since the Hubbard model in its thermodynamics limit displays a quantum phase transition at increasing U from a correlated metal to a paramagnetic Mott insulator, we would like to look at a quantity which determines a simple order parameter. Focusing on the site in the center of the small island, this is provided by the local quasiparticle residue $Z^{(0)}$ at this site 0, defined as:

$$Z^{(0)} = \frac{1}{1 + i\partial\Sigma^{(0)}(i\omega)/\partial\omega|_{\omega=0}} \quad (5)$$

introducing the local self-energy $\Sigma^{(0)}(i\omega) = i\omega + \mu - \Delta^{(0)}(i\omega) - 1/G^{(0)}(i\omega)$. For the infinite size Hubbard model, the quasiparticle weight is uniform, decreases from the value 1 at $U = 0$, and vanishes continuously at a critical strength $U_{c2}/2t \simeq 3$. For $U < U_{c2}$, strongly renormalized quasiparticles with a reduced coherence time govern the low energy properties. In parallel to the stabilization of a finite magnetization in the Ising model with a non-zero boundary magnetic field, the coupling Γ of the outer sites to the electronic environment plays the role of a dispersion, and acts in stabilizing a conducting state, even in a finite size system. The quantity $Z^{(0)}$ can thus be defined for all system sizes and interactions (even above U_{c2} provided the thermodynamic limit

is not reached). We are thus interested in the convergence of $Z^{(0)}$ with the system size L by increasing the Coulomb interaction U . This is seen on figure 3, which is obtained by using the iterated perturbation theory (IPT) method [1].

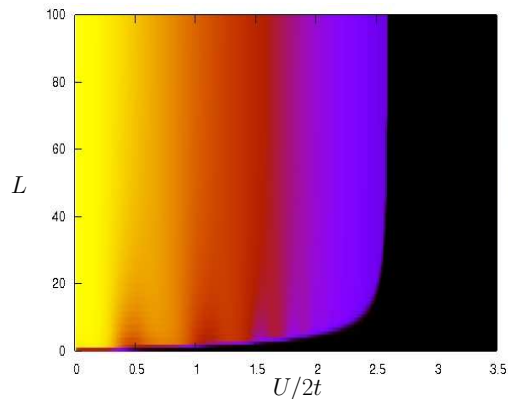


FIG. 3: Quasiparticle weight $Z^{(0)}$ in the center of the molecule as a function of Coulomb interaction $U/2t$ (abscissa) and system size L (ordinate), with external coupling to the bath $\Gamma/2t = 0.001$ and inverse temperature $\beta = 800$.

Superficially, this plot is very similar to the magnetization curve of the finite size Ising model of figure 2: for any size L , the quasiparticle weight $Z^{(0)}$ drops from 1 as U increases; also similar to the Ising case, the convergence with L towards the thermodynamic result is slow when the coupling at the boundary to the environment is small. For this precise plot, this occurs roughly for $L \sim 50$, which for an actual Bethe lattice with connectivity $z = 4$, amounts to a macroscopic system with $N = 3^{50}$ sites, an unphysically too large size, due to the fact that the Bethe lattice is very hollow. In the more realistic case of a square lattice, one obtains a number of sites $N = 100^2$, which is still very challenging for any direct numerical treatment of the complete many-body problem for such a molecule, providing a strong incentive in favor of the more economical nano-DMFT approach.

There are however some important differences if one compares closely the classical result, Fig. 2, to the quantum one, Fig. 3. First, one sees that $Z^{(0)}$ does not actually continuously vanishes at L large, but presents a small jump at $U_{c1}/(2t) \simeq 2.5$. This is due to the existence in the range $U_{c1} < U < U_{c2}$ of both metallic and insulating solutions, to either of which the system converges depending on the initial condition. For the small coupling Γ considered here (corresponding to a tiny Kondo temperature for the initial outer sites L), one starts very close to an insulating seed, explaining the discontinuous character of this transition seen in figure 3 (the metallic branch is nevertheless correctly reached for $U < U_{c1}$ and infinite DMFT iterations). One can check that the true vanishing of $Z^{(0)}$ in the thermodynamic limit at U_{c2} can be achieved by increasing the ratio $\Gamma/2t$.

More interestingly, we discover by closer inspection of figure 3 some small oscillatory behavior of the quasiparticle weight $Z^{(0)}$ as a function of U for intermediate L sizes (beyond the transient regime of very small L). To display this effect more clearly, we plot on figure 4 the relative weight to the thermodynamic limit $\delta Z^{(0)} = Z_{L \rightarrow \infty}^{(0)} - Z^{(0)}$. These oscillations constitute a very surprising result that signals that localization (at site 0) is slightly non-monotonous with increasing interaction, up to again very large system sizes. It can be

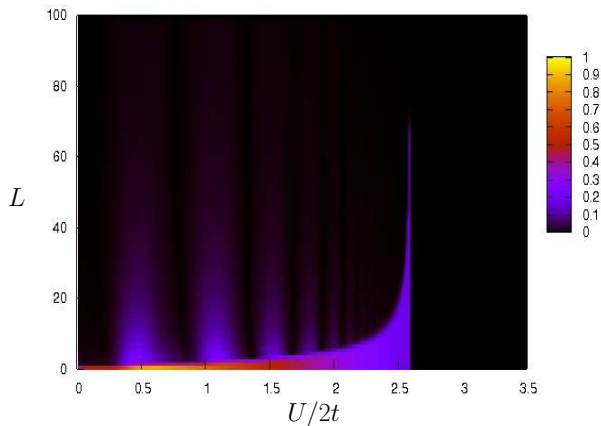


FIG. 4: Relative quasiparticle weight $\delta Z^{(0)} = Z_{L \rightarrow \infty}^{(0)} - Z^{(0)}$ for the same parameters as in figure 3.

seen also that those oscillations are displaced when comparing two adjacent sites, thus implying a non-uniform Mott localization process. Note that a similar behavior appears in the static properties of the Wigner localization in quantum dots [13]. The origin for these effects is the interplay between the interaction induced localization and the interferences within the nano object (due the presence of the boundary). This interpretation is consistent with the observation that these oscillations disappear both by increasing temperature, or by increasing the transmission at the boundary (i.e. making Γ comparable to the hopping t). This illustrates nicely how correlation effects in intermediate size nano objects may lead to unexpected and complex physical behavior.

We end the paper with some general remarks and desirable extensions of the nano-DMFT method. We would like first to emphasize that, despite some resemblances, the nano-DMFT is *not* related to the cluster-DMFT scheme [14]. Indeed, in the latter one tries to compensate the sheer locality of DMFT by exactly solving a finite size plaquette (with a total number N of sites certainly smaller than 10) embedded in a self-consistent medium that describes the effect of the remaining sites in an infinite lattice (it is thus a cavity construction with a cluster of several sites taken as cavity object). The nano-DMFT allows however to reach very large plaquette sizes, typical of big molecules, at the expense of neglecting some non-local aspects of the physics. Further

extensions of the present work should focus on investigating transport, as non homogeneities in the quasiparticle weight will also appear in the scattering rate for electron-electron interactions $\tau_{e-e}^{-1} \propto (T/Z)^2$. This asks for more sophisticated solvers for the effective action (2) than the IPT method [12], e.g. with NRG [6], and the development of a consistent formalism for computing the tunneling conductance. We stress finally that the hypothesis of a Bethe lattice is not central for the implementation of the nano-DMFT, and arbitrary “molecular” structures can be studied along the lines of Ref. [1, 15, 16]. This however needs to consider a fully iterated self-consistent nano-DMFT loop for the local Green functions, with as many local self-energies as the number of sites in the molecule.

To conclude, we have extended the DMFT of macroscopic correlated fermions models towards a nano-DMFT for generic nanoscale devices such as large molecules, quantum dot arrays, or nanostructures implanted with magnetic impurities. The method is naturally able to capture both strong electronic interactions and the coupling to an electronic environment. We have shown that these effects combine in mesoscopic systems to give interesting interplays in the Mott localization process that reveals the truly quantum nature of this phenomenon.

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